Proximity effect on the superconducting transition temperature of $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices

J. Z. Wu, C. S. Ting, and W. K. Chu
Texas Center for Superconductivity and Department of Physics, University of Houston, Houston, Texas 77204

X. X. Yao
Department of Physics, Nanjing University, Nanjing, China
(Received 20 March 1991)

The superconducting transition temperature T_c of $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices is studied theoretically with use of the Gor'kov equation. We show that the proximity effect in which the pair amplitude of the superconducting $YBa_2Cu_3O_{7-x}$ layer leaks to the semiconducting $PrBa_2Cu_3O_{7-x}$ layer plays a dominant role in explaining the depression of T_c in these superlattices with a minor modification from the charge-carrier-transfer effect across the Y and Pr interface. By adjusting the value of the hopping matrix between the Cu-O plane, our numerical result can be quantitatively compared with recent experimental measurement of T_c in $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices.

Recently, there has been considerable interest in the superconducting properties of $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices.[1-3] $YBa_2Cu_3O_{7-x}$ is a superconductor with closely spaced pairs of Cu-O planes. $PrBa_2Cu_3O_{7-x}$ has an almost identical structure to that of $YBa_2Cu_3O_{7-x}$ and is a semiconductor with very high resistivity at low temperatures. These superlattices or multilayers are stacked along the c direction as $N_Y N_{Pr} N_Y N_{Pr} N_Y N_{Pr} \cdots$. Here N_Y and N_{Pr} label, respectively, the number of Cu-O planes in $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ layers. With fixed N_Y the experimental measurements on the superconducting transition temperature T_c of the superlattices show that T_c first reduces and then saturates as N_{Pr} increases.[1,2] So far, a theoretical understanding of this problem has not been achieved, simply because the BCS equation with complicated spatial dependencies has not been solved for the superlattice even if one assumes the BCS superconductivity mechanism for $YBa_2Cu_3O_{7-x}$. In this work we try to get a semiquantitative understanding of the experimental behavior by calculating T_c of $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices for various configurations. Since the mechanism of superconductivity is not known for $YBa_2Cu_3O_{7-x}$, it will be assumed to be quasi-two-dimensional BCS like and to occur primarily in the Cu-O planes. The layers of $PrBa_2Cu_3O_{7-x}$ will be regarded as poor semiconductor with small numbers of charged carriers. Another important assumption of the present Brief Report is the existence of charge-carrier hopping between the nearest-neighboring Cu-O planes in the superlattice. Employing the Nambu's formalism,[4] a spatially dependent Gor'kov equation can be established and solved by applying the periodic boundary condition.[5] Superconducting properties of the superlattice can thus be studied. Here we shall only report our calculation on T_c and show that our result can be quantitatively compared with the experimental measurement.[1]

Our Hamiltonian is written in the Nambu's formalism[4] as follows:

$$H=\sum_{l,\mathbf{k}} \xi_l(\mathbf{k})\Psi^\dagger_{l,\mathbf{k}}\hat{\tau}_3\Psi_{l,\mathbf{k}} + \tfrac{1}{2}\sum_{l,\mathbf{k},\mathbf{k}'} V_{l,\mathbf{k},\mathbf{k}'}(\Psi^\dagger_{l,\mathbf{k}}\hat{\tau}_3\Psi_{l,\mathbf{k}'})(\Psi^\dagger_{l,-\mathbf{k}}\hat{\tau}_3\Psi_{l,-\mathbf{k}'}) + \sum_{l,l',\mathbf{k}} T_{l,l'}\Psi^\dagger_{l,\mathbf{k}}\hat{\tau}_3\Psi_{l',\mathbf{k}} , \quad (1)$$

where $\Psi_{l,\mathbf{k}}$ is the field operator, which is defined by the creation and annihilation operators of the fermion charge carriers:

$$\Psi_{l,\mathbf{k}} \equiv \begin{bmatrix} c_{l,\mathbf{k}\uparrow} \\ c^\dagger_{l,-\mathbf{k}\downarrow} \end{bmatrix} . \quad (2)$$

Here l labels the lth Cu-O plane in the $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattice. The two-dimensional wave vector $\mathbf{k}=(k_x,k_y)$ will be assumed to be continuous in the Cu-O planes. $\xi_l(\mathbf{k})\equiv\epsilon_l(\mathbf{k})-\mu_l$. μ_l and $\varepsilon_l(\mathbf{k})\equiv k^2/2m^*$ are the chemical potential and kinetic energy of carriers with effective mass m^* in the lth Cu-O plane. $V_{l,\mathbf{k},\mathbf{k}'}$, which is negative in the $YBa_2Cu_3O_{7-x}$ layers and zero in the $PrBa_2Cu_3O_{7-x}$ layers, represents the BCS pair coupling and may originate in nonphonon mechanism. $T_{l,l'}$ is the hopping matrix. $\hat{\tau}_1$, $\hat{\tau}_2$, and $\hat{\tau}_3$ are Pauli matrices. The finite-temperature Green's function is defined as

$$\hat{G}_{ll'}(\mathbf{k},\tau)\equiv -\langle \mathcal{T}_\tau[\Psi_{l,\mathbf{k}}(\tau)\Psi^\dagger_{l'k'}(0)]\rangle . \quad (3)$$

It is straightforward to show that the Green's function satisfies the Gor'kov equation based on the mean-field approximation

$$-\frac{\partial\hat{G}_{l,l'}(\mathbf{k},\tau)}{\partial\tau}=\delta_{ll'}\delta(\tau)+[\xi_l(\mathbf{k})\hat{\tau}_3+\Delta_l(\mathbf{k})\hat{\tau}_1]\hat{G}_{l,l'}(\mathbf{k},\tau) + \sum_m T_{l,m}\hat{\tau}_3\hat{G}_{m,l'}(\mathbf{k},\tau) . \quad (4)$$

Here the gap function or the order parameter Δ_l is defined as

$$\Delta_l = -V_l \sum_{\mathbf{k}} F_{l,l}(\mathbf{k},0) , \tag{5}$$

and we have used the s-wave approximation for the pair interaction

$$V_{l,\mathbf{k},\mathbf{k}'} = \begin{cases} -V_l, & |\xi_l(\mathbf{k})| < E_f(l) \\ 0, & \text{otherwise} . \end{cases} \tag{6}$$

$F_{l,l}(\mathbf{k},0) = -\langle \mathcal{T}_\tau [c_{l,\mathbf{k}\uparrow} c_{l,-\mathbf{k}\downarrow}] \rangle$ is the off-diagonal matrix element of $\hat{G}_{l,l}(\mathbf{k},\tau)$ at $\tau=0$. Introducing Matsubara's Fourier transform,[6] Eq. (4) can be reduced to the following matrix equation:

$$\sum_m \hat{K}_{l,m} \hat{G}_{m,l'}(\mathbf{k},\omega_n) = \hat{\delta}_{l,l'} , \tag{7}$$

where the matrix $\hat{K}_{l,l'}$ is defined as

$$\hat{K}_{l,l} \equiv i\omega_n \hat{I} - \xi_l(\mathbf{k})\hat{\tau}_3 - \Delta_l(\mathbf{k})\hat{\tau}_1 , \tag{8}$$

$$\hat{K}_{l,l'} \equiv -T_{ll'}\hat{\tau}_3, \quad l \neq l' . \tag{9}$$

$\omega_n \equiv (2n+1)\pi k_B T$ is the Matsubara's frequency for fermions. Considering that the hopping matrix $T_{l,l'}$ is weak and short ranged, we may assume that only the charge-carrier hopping between the nearest-neighbor Cu-O planes is important. Under this condition, the hopping matrix $T_{l,l'}$ becomes

$$T_{l,l'} = \begin{cases} J_1 \delta_{l,l\pm1}, & \text{inside } YBa_2Cu_3O_{7-x} \\ J_2 \delta_{l,l\pm1}, & \text{inside } PrBa_2Cu_3O_{7-x} \\ J_3 \delta_{l,l\pm1}, & \text{Y and Pr interface} . \end{cases} \tag{10}$$

Employing the above equation, the matrix $\hat{K}_{ll'}$ defined in Eqs. (8) and (9) reduces to a tridiagonal matrix with each element being a 2×2 matrix. However, Eq. (7) is still very difficult to solve because the dimension of the matrix $\hat{K}$ with $\hat{K}_{l,l'}$ as its element equals to the number of Cu-O planes in the superlattices. Since the superlattice is a periodic system with enlarged unit-cell size $N = N_Y + N_{Pr}$, we may apply the periodic boundary condition for the hopping matrix, which is $T_{N,N+1} = T_{N,1}$. Therefore, $\hat{K}_{ll'}$ is simplified to an $N\times N$ matrix. By extending the method developed by Dy, Wu, and Spratlin,[7] we are able to solve Eq. (7) by summing up all the Feynman diagrams under the periodic boundary conditions for the superlattice. With the solution of Eq. (7), the local pair amplitude in each Cu-O plane can be determined self-consistently by the following equation:

$$F_{l,l}(0,0) = 2N_l k_B T \int_0^{E_f(l)} d\xi_l \sum_{n=-\infty}^{\infty} F_{l,l}(\mathbf{k},\omega_n) ,$$

$$l = 1,2,\ldots,N . \tag{11}$$

Here k_B is the Boltzmann constant. N_l is the density of state for the charge carriers in the lth Cu-O plane. It is expected that all the charge carriers in $YBa_2Cu_3O_{7-x}$ contribute to the superconductivity; the cutoff energy in Eq. (11) is therefore chosen to be the Fermi energy $E_F(l)$. Equation (11) can then be employed to study the proximity effect or how the superconducting pairs leak from the $YBa_2Cu_3O_{7-x}$ layer to the $PrBa_2Cu_3O_{7-x}$ layer. The expression of $F_{l,l}(\mathbf{k},\omega_n)$ is rather lengthy and will not be listed here. However, a detailed derivations of $F_{l,l}(\mathbf{k},\omega_n)$ together with $\hat{G}_{l,l}(\mathbf{k},\tau)$ is going to be presented elsewhere for publication.[5] $E_f(l) = \mu_l$ at zero temperature could vary drastically for different Cu-O planes because of the charge-carrier density in the $PrBa_2Cu_3O_{7-x}$ being usually found 2 to 3 orders smaller than that of the high-T_c $YBa_2Cu_3O_{7-x}$ superconductor.[8] This big difference in the charge-carrier densities may result in a substantial charge-carrier transfer across the $YBa_2Cu_3O_{7-x}$/$PrBa_2Cu_3O_{7-x}$ interface, which in turn could affect the Fermi energy after redistribution of the charge carriers. In order to study this effect, we adopt a semiclassical approach. The change of the charge-carrier density in the lth Cu-O plane is

$$\delta n(l) = \int_{U(l)}^{E_F} N_l dE - \int_0^{E_F(l)} N_l dE , \tag{12}$$

where $E_F(l)$ and E_f are the Fermi energy before and after the charge-carrier transfer in the lth Cu-O plane. $U(l)$, the electrostatic potential shift due to $\delta n(l)$, can be solved by the Poisson equation in the form of a difference equation,

$$U(l+1) - 2U(l) + U(l-1) - \frac{4\pi e^2 d^2 \delta n(l)}{\epsilon(l)} = 0 , \tag{13}$$

self-consistently in combination with Eq. (12). Here the condition of the total number of the charge carriers being unchanged during the transfer process is rigorously imposed. We have also assumed that the electrostatic potentials for $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ are both equal to zero before the charge transfer. d is the interlayer spacing, which is estimated to be ~ 6 Å for $YBa_2Cu_3O_{7-x}$ as well as for $PrBa_2Cu_3O_{7-x}$. The dielectric constant $\epsilon(l)$ is assumed to be 10 for both $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ based on the experimental result.[9] Our calculation has indicated that the charge-carrier transfers are limited within one and two to three Cu-O layers, respectively, on the $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ sides from the interface. The relative change in the Cu-O plane of the superconducting side is about 1%, while the change of the charge-carrier density could be as large as 100–150 % in the Cu-O plane of semiconducting side. Since the superconducting properties of the $YBa_2Cu_3O_{7-x}$/$PrBa_2Cu_3O_{7-x}$ multilayer is mostly determined by the $YBa_2Cu_3O_{7-x}$ layers, the effect of the charge-carrier transfer would become important when the thickness of the $YBa_2Cu_3O_{7-x}$ layer comprises only one unit cell or two Cu-O planes. Considering the charge-carrier transfer, the integration range in Eq. (11) is shifted from $(0,E_f(l))$ to $(0,E_f - U(l))$ for the lth layer. In our calculations the Fermi energies before the charge-carrier transfer are chosen to be 0.127 and 0.028 eV, respectively, for $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$. By doing so, the effective mass of $PrBa_2Cu_3O_{7-x}$ is assumed to be much smaller than that of $YBa_2Cu_3O_{7-x}$. The charge-carrier densities are chosen to be

$4.0\times10^{21}/cm^3$ for $YBa_2Cu_3O_{7-x}$ and $1.0\times10^{19}/cm^3$ for $PrBa_2Cu_3O_{7-x}$. V_lN_l for $YBa_2Cu_3O_{7-x}$ is taken as 0.339 to give a T_c about 90 K based on BCS theory. By plotting $F_{l,l}(0,0)$ against the temperature T, T_c can be obtained numerically by requiring $F_{l,l}(0,0)=0$ at $T=T_c$.

In Fig. 1 we have plotted the transition temperature of the superlattice as a function of the number of Cu-O planes N_{Pr} in a $PrBa_2Cu_3O_{7-x}$ layer for the hopping constant $J_i=0.03$ eV ($i=1,2,3$). $N_Y=2,4,6,8$ represents the number of Cu-O planes in a $YBa_2Cu_3O_{7-x}$ layer. T_{c0} is the transition temperature for $YBa_2Cu_3O_{7-x}$ film, while it is thick enough so that the surface or interface effect becomes unimportant. In our calculation both the charge-carrier-transfer and proximity effects are considered. It is shown in Fig. 1 that as the thickness or N_{Pr} of the $PrBa_2Cu_3O_{7-x}$ increases, the transition temperature of the superlattice initially drops drastically and then approaches to a saturated value T_s for large N_{Pr}. This phenomenon has been observed in experiments by Lowndes *et al.*[1] and Li *et al.*,[2] respectively. The measured T_s are 19, 54, 71, and 80 K approximately for $N_Y=2$, 4, 6, and 8, which gives $T_s/T_{c0}=0.2$, 0.58, 0.76, and 0.86 at $N_{Pr}=32$ with $T_{c0}\sim92.5$ K (asterisks in Fig. 1). Here N_{Pr} related with the thickness d_{Pr} of the $PrBa_2Cu_3O_{7-x}$ layer through the relation $d_{Pr}=(N_{Pr}/2)c$ with $c=2d$. Our calculation gives the corresponding values of T_s/T_{c0} as 0.19, 0.58, 0.80, and 0.88, which agree well with the experimental observation.[1] In order to fit the experimental measurement, we have chosen $J_1=J_2=J_3$. Since the lattice structures of $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ are almost identical, the choice of $J_1=J_2=J_3$ should be reasonable and it reduces the number of parameters. However, the values of J_i do influence our result. For example, if the value of $J_i=0.02$ eV ($i=1,2,3$) is taken, our numerical result would show a similar behavior, but the saturated transition temperature T_s becomes higher. In other words, T_s increases as the value of J_i decreases. A physical explanation of the experimental measured behavior of T_c will be presented later.

In Fig. 2 the dependence of T_c on N_Y or the thickness of the $YBa_2Cu_3O_{7-x}$ layer is shown for $N_{pr}=4$, 8, and 32. Our numerical results are also in good agreement with the experimental measurements.[1] It is very interesting to note that the $YBa_2Cu_3O_{7-x}$ layer, as thin as only one unit cell ($N_Y=2$), still has a transition temperature of about 19 K, while the $PrBa_2Cu_3O_{7-x}$ layer is 10–15 times thicker. Thus it confirms our assumption that the intralayer pairing is a dominant contribution to the superconductivity in $YBa_2Cu_3O_{7-x}$. On the other hand, experimental measurements for ultrathin $YBa_2Cu_3O_{7-x}$ films[10] have shown that no superconductivity could sustain in one- or two-unit-cell-thick film. This result could be attributed to the existence of large distortions in the lattice structure of $YBa_2Cu_3O_{7-x}$ film near the free surface and interface with the substrate. In the $YBa_2Cu_3O_{7-x}$/ $PrBa_2Cu_3O_{7-x}$ superlattices, the layers of $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ are well matched and there exist no distortions near the interfaces. This is the reason why a finite transition temperature can still be sustainable even for the case of $N_Y=2$. However, T_c is still much depressed in a superlattice with a very thin $YBa_2Cu_3O_{7-x}$ layer because both the charge-carrier-transfer and proximity effects become more pronounced

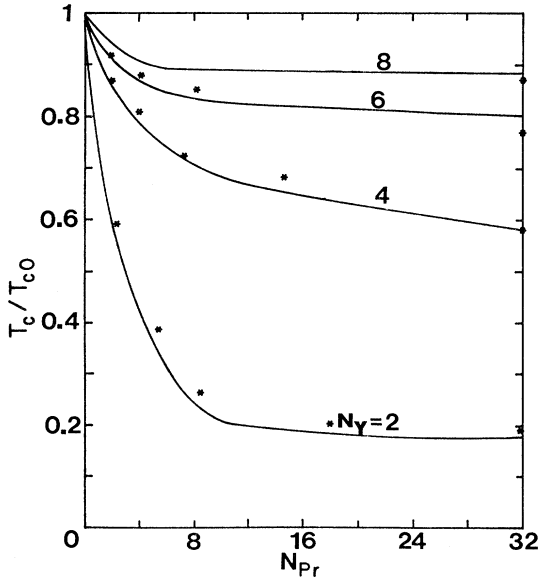

FIG. 1. Normalized transition temperature T_c/T_{c0} of $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices vs N_{Pr} for $J_i=0.03$ eV ($i=1,2,3$). The asterisks represent the experimentally measured data in Ref. 1.

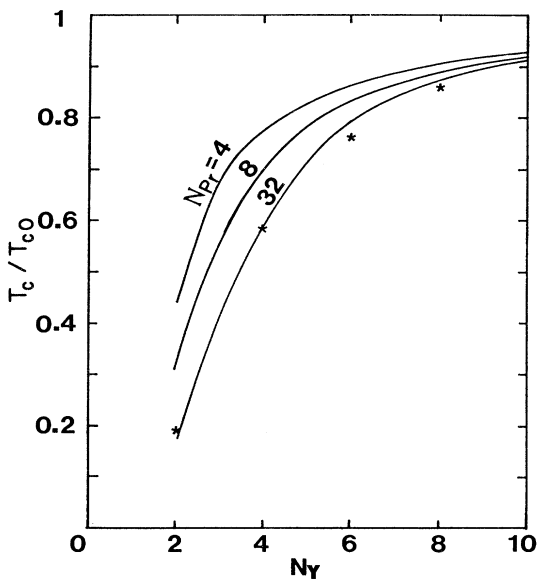

FIG. 2. Normalized transition temperature T_c/T_{c0} of $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices vs N_Y for $J_i=0.03$ eV ($i=1,2,3$) and $N_{Pr}=4,8$ and 32. The asterisks represent the experimental data taken from Ref. 1 for $N_{Pr}\sim32$.

there.

To understand the proximity effect and the degree of the pair amplitude leaking from the superconducting layer to the semiconducting layer, we have plotted in Fig. 3 the change of the normalized pair amplitude across the $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ interface for $T/T_{c0}=0.63$, $J_i=0.03$ eV, $N_Y=4$, and $N_{Pr}=8$. F^* is the pair amplitude in layer 1. We have noted that even though the pair amplitude inside $PrBa_2Cu_3O_{7-x}$ is not zero, its magnitude decreased exponentially from the interface. The Cooper pair leaking length ξ_N, which is calculated from $F_{l,l}(0,0)\sim\exp[-(l-l_0)d/\xi_N]$ in $PrBa_2Cu_3O_{7-x}$ with $l_0=3$, is about the length of 2.5 Cu-O planes under the above parameters. That implies that the proximity effect is localized within range ξ_N at the interface. Here we wish to emphasize that the superconducting order parameter is different from the pair amplitude. The order parameter in $PrBa_2Cu_3O_{7-x}$ is zero because of the BCS pair interaction being assumed to be zero there. Down to the deep layer of the $PrBa_2Cu_3O_{7-x}$, there is little pair amplitude when d_{Pr} is more than 2 or 3 times larger than ξ_N so that the superconducting phase is actually confined within $YBa_2Cu_3O_{7-x}$ layers.

In summary, the transition temperature T_c of the $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices has been studied theoretically by solving the spatially dependent Gor'kov equation. With fixed N_Y, our numerical calculation shows that T_c decreases and then saturates as d_{Pr} is increased. With fixed N_{Pr} or d_{Pr}, T_c is found to be reduced as N_Y is decreased. One of the essential features in our result is that T_c is still finite even for $N_Y=2$ (the $YBa_2Cu_3O_{7-x}$ layer contains a single unit cell) and large N_{Pr}. This result is consistent with experimental measurements and shows that the superconductivity originated primarily in the Cu-O planes. The decrease of T_c is caused by the charge-carrier transfer effect near the interface and most importantly by the proximity effect in which the pair amplitude in the $YBa_2Cu_3O_{7-x}$ layer leaks to the $PrBa_2Cu_3O_{7-x}$ layer. Since these effects are confined only within two or three Cu-O planes on the $PrBa_2Cu_3O_{7-x}$ side of the interface, further increasing N_{Pr} or d_{Pr} has no significant influence on T_c. This is the reason why T_c first reduces and then saturates as the value of N_{Pr} is increased. Finally, we wish to emphasize that our result depends on the existence of a hopping matrix element between the nearest-neighbor Cu-O planes so that the charge-transfer and proximity effects could be both present, not very much on whether the $PrBa_2Cu_3O_{7-x}$ layer is a semiconductor or an insulator with its charge carrier being localized.

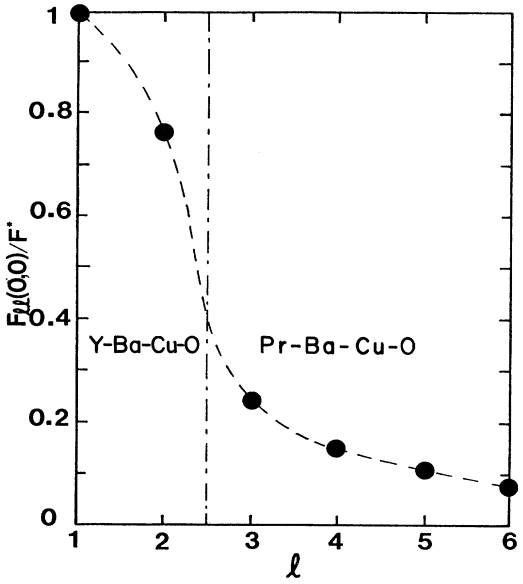

FIG. 3. Normalized pair amplitude across the $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ interface with $N_Y=4$, $N_{Pr}=8$, $T/T_{c0}=0.63$, and $J_i=0.03$ eV ($i=1,2,3$). The solid circles represent the results of numerical calculation and l is the layer label.

We would like to thank S. J. Xiong for helpful discussion. This work is supported by the Texas Center for Superconductivity at University of Houston under Grant No. MDA 972-8-G-0002 to the University of Houston from DARPA, the State of Texas, and also a grant from the Robert A. Welch foundation.

[1]D. H. Lowndes *et al.*, Phys. Rev. Lett. **65**, 1160 (1990).
[2]Q. Li *et al.*, Phys. Rev. Lett. **65**, 3086 (1990).
[3]J. M. Triscone *et al.*, Phys. Rev. Lett. **64**, 804 (1990).
[4]J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1983).
[5]J. Z. Wu, X. Yao, and C. S. Ting (unpublished).
[6]G. D. Mahan, *Many-Particle Physics* (Plenum, New York, 1981).
[7]K. S. Dy, S. Y. Wu, and T. Spratlin, Phys. Rev. B **20**, 4237 (1979).
[8]X. D. Wu *et al.*, Appl. Phys. Lett. **56**, 400 (1990).
[9]S. Massidda *et al.*, Phys. Lett. A **122**, 198 (1987); B. Renker *et al.*, Jpn. J. Appl. Phys. **26**, Suppl. 3, 2134 (1987); G. A. Samara *et al.*, Phys. Rev. Rev. B **41**, 8974 (1990).
[10]X. X. Xing *et al.*, Appl. Phys. Lett. **54**, 2376 (1989); A. F. Hebard *et al.*, *ibid.* **55**, 1945 (1989).